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ON THE ZERO DIFFUSIVITY INVARIANCE
FOR CHEMICAL REACTION IN
A TURBULENT FLUID

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CHEMICAL REACTION IN A TURBULENT FLUID

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ABSTRACT

It is suggested that the distribution function approach be used for describing the turbulent chemical reaction in order to avoid violation of an invariance property noted by O'Brien: for zero molecular diffusion in homogeneous turbulence, single point statistical functions of the concentration field are independent of the motion. This property and a similar "white noise" invariance for two point statistical functions are shown to apply to arbitrary isochoric motions when the initial concentration field is statistically homogeneous with respect to position. Finally, it is observed that the use of a reciprocal concentration avoids the problem of preserving the invariance in turbulence closure schemes.

I. INTRODUCTION

O'Brien^{1,2} has recently noted an invariance property of the model second order irreversible chemical reaction in turbulent flow³

$$\mathcal{N}(x, t) \Gamma(x, t) = -C \Gamma^2(x, t) \quad (1)$$

where $\mathcal{N}(x, t) = \{\partial/\partial t + U(x, t) \cdot \nabla - D \nabla^2\}$, Γ is the mass concentration in either mass or molar units (with appropriate dimensions of C), $U(x, t)$ is the incompressible mass-average velocity field, D is the constant molecular diffusivity of the species Γ with respect to the mixture, and C is a constant reaction rate constant. The restrictions that U be unaffected by Γ or by temperature changes and that D and C be constant usually require low Γ and either small heats of reaction or large heat capacities. That no reverse reaction or creation term appears in (1) usually restricts one to the initial stages of reaction, unless Γ is extremely dilute in the product of reaction. We adopt the outlook that Γ is a passive contaminant.

The invariance property in homogeneous turbulence is that for $D = 0$ all statistical functions $\langle \Gamma^n(x, t) \rangle$ $n = 1, 2, \dots$ are independent of the function U . Here the angular brackets denote ensemble average over all realizations of U . In homogeneous turbulence we require (in this paper) that statistical functions of U and Γ are independent of position x . In the more general case, when U is

an arbitrary isochoric motion with no flow across the boundaries, the invariance requires that for $D = 0$ all volume averages of $\Gamma^n(x, t)$ be independent of U . We will not discuss the latter case here.

One can show the invariance directly from (1) by setting up an unclosed hierarchy of statistical moment equations in homogeneous turbulence (or of volume averages in the general case). This hierarchy is independent of U . Alternatively, in Lagrangian coordinates the fluid motion enters (1) only through the Lagrangian form of the diffusion term. For $D = 0$ the behavior of Γ on any trajectory or particle path is independent of U , and the invariance follows from the requirement that Γ is initially statistically homogeneous, the identity of Lagrangian and Eulerian volume averages in isochoric motion, and an ergodic assumption on the identity of statistical and volume averages. Later, this last assumption will be relaxed.

The purpose of this paper is to see how the invariance may be preserved in closure theories. We will present arguments for the distribution function approach and look at the one and two point probability density functions of Γ for $D = 0$. Then we will show that a simple change of variables can remove the problem.

II. PRESERVING THE INVARIANCE

There are two types of dynamic nonlinearity of (1) leading to closure difficulty of the moment equations. First, the nonlinearity of the reaction term and

statistical initial and boundary conditions produce coupling between moments of all orders of Γ . This difficulty is formally the same as that of the turbulence problem itself (closure of the Navier-Stokes equations), except that the coupling in (1) is local in x -space. Second, randomness is induced by the turbulence, analogous to the nonreactive passive scalar problem. Of course, the nonlinearities are not independent, as the coupling in the reaction term will be induced by turbulence alone if the initial and boundary conditions of Γ are certain.

Many of the analytical methods in turbulence and turbulent convection theory have been treated as procedures for forming moment closures regardless of the nature of the dynamic nonlinearity. The direct interaction (DI) approximation⁴ and the Lagrangian history direct interaction (LHDI) approximation,⁵ as they have been applied to (1),^{2,6,7} clearly fall in this category, as do the more classical approaches of iteration expansions and cumulant discards.^{1,8} The phenomenological theories have not been considered for this problem, since the reaction term is in no sense a transport term, but rather is purely dissipative.

If the moment closure does not explicitly involve the velocity field in the reaction term, such as an ad hoc cumulant discard hypothesis applied to moments of Γ , the zero diffusion invariance will not be violated, although the decay rates of the moments of Γ and the influence of the velocity field for $D \neq 0$ may be given incorrectly. The DI approximation applied to (1)⁶ does involve the velocity field in the reaction term but fails to preserve the invariance.² The

reaction is not treated exactly, as the model for which the DI approximation is the exact solution in the reaction problem involves artificial dynamical coupling in both the convection and reaction terms. However, the LHDI approximation (and the abridged version) does preserve the invariance.⁷ Its success can be traced to the fact that the averaged response function G for infinitesimal concentration perturbations retains two important properties of the Lagrangian form of (1) for $D = 0$: first, all fluid particle trajectories are independent (G is non-zero only for perturbation and response on the same trajectory), and second, all trajectories behave identically (G is independent of labelling time) so that G is independent of the velocity field. Despite this success, there is no guarantee of the accuracy of the LHDI's treatment of the reaction term. Furthermore, because of the presence of Lagrangian velocity correlations, we regard the LHDI to be too difficult in practice.

If one takes the point of view⁹ that the turbulence, acting through the random coefficient U , should provide the only stochastic indeterminacy in (1), and that the effect of the reaction should be treated exactly, as the diffusion term is customarily done, then any such closure method should retain the zero diffusivity invariance. Actually, the reaction need be treated exactly only in the limit $D = 0$.

It is clear that if the reaction term is to be treated exactly, the moment closure approach is not appropriate to the study of (1). For example, any iteration procedure based upon treating the turbulent velocity as a perturbation

is very difficult in that it requires solution of a nonlinear and nonhomogeneous partial differential equation at each level of iteration. Even for $D \approx 0$ one is confronted with a sequence of Riccati equations for Γ .

On the other hand, the distribution function approach seems appropriate because the reaction term causes no extra conceptual difficulty above that of the nonreactive passive scalar problem. The closure problem here is not due to the nonlinearity of the dynamical equation, but to nonlocalness of the dynamical variables. The reaction term is completely local in x -space, whereas the diffusion and convection terms are not.

In the next sections we briefly outline the distribution function approach and examine the zero diffusion invariance in greater detail.

III. THE DISTRIBUTION FUNCTION APPROACH

We now sketch out the distribution function approach to justify our preceding remarks and to focus on its closure difficulty. The overall objective is to obtain at time t the one and two point probability density functions (pdf's) of Γ defined by

$$P_x^t(\psi) d\psi = P \{ \psi \leq \Gamma(x, t) < \psi + d\psi \} \quad (2)$$

$$P_{x_1, x_2}^t(\psi_1, \psi_2) d\psi_1 d\psi_2 = P \{ \psi_1 \leq \Gamma(x_1, t) < \psi_1 + d\psi_1, \psi_2 \leq \Gamma(x_2, t) < \psi_2 + d\psi_2 \}$$

where P denotes probability. These pdf's will allow us to calculate mean decay rates and two point correlations of Γ and may be obtained by reduction from the full distribution functions. Let $P^t [\psi(x) | u(y)]$, or $P^t [\psi | u]$, denote the conditional probability functional at time t for the concentration field Γ , given the velocity field U , i.e.

$$P^t [\psi(x) | u(y)] \delta\psi(x) = P \{ \psi(x) \leq \Gamma(x, t) < \psi(x) + d\psi(x), \forall x |$$

$$u(y) \leq U(y, t) < u(y) + du(y), \forall y \}$$

where the conditional events are to the right of the vertical bar, and $\delta\psi$ is elemental volume in ψ -space. Strictly speaking, the probability density functional does not exist in infinite dimensional phase space where $\delta\psi$ cannot be defined, and must be interpreted as the joint pdf of Γ at an arbitrarily large number of points x_i . A Liouville or continuity equation for $P^t [\psi | u]$ may be derived from (1) with the method used by Edwards¹⁰; we get

$$-\frac{\partial}{\partial t} P^t [\psi | u] = \int d^3x \frac{\delta}{\delta\psi(x)} [C\psi^2(x) + u(x) \cdot \nabla\psi(x) - D\nabla^2\psi(x)] P^t [\psi | u] \quad (3)$$

where δ / δ denotes functional differentiation.

To obtain an equation for $P^t [\psi]$, the marginal functional distribution, multiply (3) by the full pdf of U for all time, i.e. by $F[u]$, defined by

$$F[u] \delta v = P\{v(y, s) \leq U(y, s) < v(y, s) + dv(y, s), \forall y \text{ and } 0 \leq s \leq T\}$$

where T is some finite time greater than any considered in the problem, and then perform a functional integration over all v . The result is similar to (3) except for an integral over the convection term involving the joint distribution of ψ with u . The ability to evaluate this term at any t is tantamount to knowing $F[u]$, whereas in practice we can only deal with a few moments of u . The procedure of Ref. 9 is to expand $P^t[\psi | u]$ in the convection term about $P^t[\psi]$ before carrying out the v integration, in order to obtain an expression for $P^t[\psi]$ in terms of moments of v .

The single point pdf is obtained by multiplying (3) by $F[u]$ and integrating over all v and over ψ for all points except x . The result is

$$\left[\frac{\partial}{\partial t} - C \frac{\partial}{\partial \psi} \psi^2 \right] P_x^t(\psi) = - D \lim_{x' \rightarrow x} \frac{\partial}{\partial \psi} \nabla_{x'}^2 \int d\psi' \psi' P_{x,x'}^t(\psi, \psi') - \int d^3 u u \cdot \nabla P_{x,x}^t(\psi, u) \quad (4)$$

where the convection term, which involves the joint distribution of ψ with u , must be approximated, say as in the preceding paragraph. For the particular case of joint statistical homogeneity of Γ and U , i.e. $P_{x,x}^t(\psi, u)$ is independent

of \mathbf{x} , the convection term in (4) is zero. If D were also zero, then $P^t(\psi)$ may be calculated from the result for zero motion in accordance with O'Brien's invariance. We shall discuss this in more detail later. Unfortunately, there is also a closure problem associated with the nonlocalness of the diffusion term. An unclosed hierarchy for these multipoint pdf's can be set up, with each equation similar in form to (4), and with the reaction term treated exactly, as on the left hand side of (4). For example the two point pdf obeys the equation

$$\left[\frac{\partial}{\partial t} - C \left(\frac{\partial}{\partial \psi_1} \psi_1^2 + \frac{\partial}{\partial \psi_2} \psi_2^2 \right) \right] P_{x_1, x_2}^t (\psi_1, \psi_2) =$$

$$- D \left\{ \lim_{x_3 \rightarrow x_1} \frac{\partial}{\partial \psi_1} + \lim_{x_3 \rightarrow x_2} \frac{\partial}{\partial \psi_2} \right\} \nabla_{x_3}^2 \int d\psi_3 \psi_3 P_{x_1, x_2, x_3}^t (\psi_1, \psi_2, \psi_3)$$

$$- \int d^3 u u \cdot \left[\nabla_{x_1} P_{x_1, x_2, x_1}^t (\psi_1, \psi_2, u) + \nabla_{x_2} P_{x_1, x_2, x_2}^t (\psi_1, \psi_2, u) \right].$$

Higher order equations may be obtained by induction. This sequence is analogous to that for the ordinary turbulence problem.^{11, 12}

In the ordinary passive scalar problem, for which $C = 0$, the nonlocalness of the diffusion term could be removed by using the spatial Fourier transform of (1). But for $C \neq 0$ the reaction term in wavenumber coordinates is a convolution, and closure on that term may lead to violation of the $D = 0$ invariance. In

x -coordinates a closure must be made on the diffusion term, but the reaction may be treated exactly and will preserve the invariance. However, for small C , large D , or in regions of phase space where $|\nabla\psi|$ is large, the invariance may not be as important, and in those instances it is more important that the diffusion be treated exactly.

We suggest the following method for dealing with the diffusion term. First, for the velocity term, make a suitable closure for $P^t[\psi | u]$. Then, base the reduction from $P^t[\psi]$ for use in the diffusion term, say at the two point level, upon the formulation of (1) in k -space, where the diffusion may be treated exactly at the expense of exact treatment of the reaction. This approach would guarantee correct handling of the reaction term in (4) when diffusion is weak, with the diffusion term being treated properly when diffusion is dominant.

IV. THE SINGLE POINT PROBABILITY DENSITY FUNCTION FOR $D = 0$

Let us now consider the single point probability density function (pdf) of Γ in the limit $D = 0$ to see what form must be retained in any distribution function approach that satisfies O'Brien's invariance. In this section, the term homogeneity means that the attribute in question, say $\Gamma(x, t)$, is statistically homogeneous with respect to position: the probability density function of the attribute is independent of the position of the attribute.

First, examine the case when there is no motion, $U(x, t) = 0$. Then (1) reduces to

$$\frac{\partial}{\partial t} \Gamma(x, t) + C \Gamma^2(x, t) = 0 \quad (5)$$

which has already been studied by O'Brien.¹ The solution of (5) is

$$\Gamma(x, t) \equiv 2^t \Gamma(x, 0) = \Gamma(x, 0) [1 + Ct \Gamma(x, 0)]^{-1}.$$

The single point pdf of $\Gamma(x, t)$ can be related to the initial distribution of Γ by the identity

$$P_x^t(\psi) = P_x^0(2^{-t}\psi) \left| \frac{d2^{-t}\psi}{d\psi} \right| \quad (6)$$

Now introduce the condition that Γ is initially homogeneous, i.e. P^0 is independent of x . From (6) the same condition prevails for Γ at time t , whereby

$$P^t(\psi) = (1 - Ct\psi)^{-2} P^0 \left(\frac{\psi}{1 - Ct\psi} \right). \quad (7)$$

We now observe that P^t is the same as (7) for $D = 0$ in homogeneous turbulence if P^0 is independent of x . This is clear from (4). Alternatively, the single point moment equations, which are independent of U in accordance with

O'Brien's invariance, are the same as those for (5); and because the pdf, or equivalently the characteristic function, is determined completely by the moments, the equality of the P's follows.

Let us examine P^t from another point of view. Rewrite (1) in Lagrangian form as

$$\frac{\partial}{\partial t} \tilde{\Gamma}(\xi, t) + C \tilde{\Gamma}^2(\xi, t) = D C^{-1}(\xi, t) : \nabla_{\xi} \nabla_{\xi} \tilde{\Gamma}(\xi, t) \quad (8)$$

where ξ is position in the Lagrangian or material frame, C^{-1} is the inverse Green deformation tensor

$$C^{-1}(\xi, t) = [(\nabla f^{-1}) \cdot (\nabla f^{-1})^{tr}]_{x=f(\xi, t)},$$

the motion is given by $x = f(\xi, t)$ with inverse $\xi = f^{-1}(x, t)$ and velocity $U(x, t) = \partial f(\xi, t)/\partial t$, the Eulerian and Lagrangian concentrations are related as

$$\Gamma(x, t) = \Gamma(f(\xi, t), t) \equiv \tilde{\Gamma}(\xi, t),$$

and ∇ and ∇_{ξ} are the Eulerian and Lagrangian del operators.¹³ In analogy with (2) we introduce the Lagrangian single particle pdf of Γ

$$\tilde{P}_{\xi}^t(\psi) d\psi = P\{\psi \leq \tilde{\Gamma}(\xi, t) < \psi + d\psi\},$$

also the conditional Eulerian pdf of Γ for a particular fluid particle

$$P_x^t(\psi) d\psi = P \{ \psi \leq \Gamma(x, t) < \psi + d\psi \mid g \leq f^{-1}(x, t) < g + dg \}$$

and the pdf for the initialization of trajectories that pass through x at the time t

$$F_x^t(g) d^3g = P \{ g \leq f^{-1}(x, t) < g + dg \}.$$

For the case $D = 0$ (8) has the same form as (5), i.e.

$$\frac{\partial}{\partial t} \tilde{\Gamma}(\xi, t) + C \tilde{\Gamma}^2(\xi, t) = 0, \quad (9)$$

and for homogeneous initial conditions $\tilde{P}_\xi^0 = P_x^0 = P^0$ independent of x or ξ ,

where we have noted $f(\xi, 0) = \xi$. From (6) we obtain the result analogous to (7)

$$\tilde{P}_\xi^t(\psi) = (1 - Ct\psi)^{-2} P^0 \left(\frac{\psi}{1 - Ct\psi} \right) = \tilde{P}^t(\psi) \quad (10)$$

independent of ξ . The Eulerian pdf is recovered by summing the conditional pdf over all particles,

$$P_x^t(\psi) = \int P_x^t|_g(\psi) F_x^t(g) d^3g. \quad (11)$$

Note that $P_x^t|_g(\psi)$ and $\tilde{P}_g^t(\psi)$ are pdf's for the same fluid particle g at the same time; they are therefore equal and, from (10), independent of g . Consequently, g integrates out of (11) with the result

$$P_x^t(\psi) = \tilde{P}^t(\psi)$$

independent of x and of the motion, or

$$P^t(\psi) = \tilde{P}^t(\psi) = (1 - Ct\psi)^{-2} P^0 \left(\frac{\psi}{1 - Ct\psi} \right). \quad (12)$$

Recall the weakness of the assumptions used in obtaining (12). Of course $D = 0$, $C = \text{constant}$, and $P^0(\psi)$ must be independent of x . The motion must also be isochoric so that (9) does not contain a dilatation term. However, we have made no assumption about the turbulence itself, such as spatial homogeneity of the velocity fluid. The derivation does not even require the motion to be turbulent, as (12) holds for each realization of the motion. We have had to introduce, however, an ensemble of initial Γ fields in addition to the ensemble of velocity fields.

The following physical picture is presented by (12). Because \tilde{P}^0 is the same for all particles, any particle that passes x at time t had the same \tilde{P}^0 in each realization of U and also the same age t . Because \tilde{P}^t depends only on age for $D = 0$, it will be the same at x in each realization regardless of the motion, and the same for all points x .

Our conclusion for $D = 0$ is, then, that if at some time Γ becomes homogeneous, it will remain so independent of the subsequent motion, and P^t may be calculated from the result (7) for zero motion. It would also be desirable to determine the circumstances under which Γ becomes homogeneous, although we will not investigate this here. (We would like to use the continuity equation for $P^t[\psi|u]$ to show that Γ tends toward homogeneity if the U field is completely homogeneous.)

The Lagrangian formulation had to be used here because there is no counterpart to (6) in the Eulerian formulation. The order of the reaction is unimportant; in other instances the equivalent of (9) would be that $\tilde{\Gamma}(\xi, t)$ be independent of other fluid particles so that (6) can be used. For no reaction we obtain the familiar passive scalar result $P^t(\psi) = P^0(\psi)$, i.e. all moments of Γ are constants of the motion.

V. A "WHITE NOISE" INVARIANCE FOR $D = 0$

In addition to the property of the single point pdf given by (12) for $D = 0$, there is a similar invariance for the two point pdf of Γ : if Γ is initially homogeneous and pairwise independent for all points, then it will remain so regardless of the subsequent motion.

The proof is similar to that for (12), except for the use of $P_{x_1, x_2}^t(\psi_1, \psi_2)$, the two point pdf of Γ ; $\tilde{P}_{\xi_1, \xi_2}^t(\psi_1, \psi_2)$, the two particle pdf of Γ ; $P_{x_1, x_2}^t|_{g_1, g_2}(\psi_1, \psi_2)$, the conditional Eulerian pdf of Γ for two particles; and $F_{x_1, x_2}^t(g_1, g_2)$,

the joint pdf of particles passing through x_1 and x_2 at time t - all defined by analogy to the single point functions. The formula equivalent to (11) is

$$\begin{aligned} P_{x_1, x_2}^t(\psi_1, \psi_2) &= \iint P_{x_1, x_2}^t|_{\xi_1, \xi_2}(\psi_1, \psi_2) F_{x_1, x_2}^t(g_1, g_2) d^3g_1 d^3g_2 \\ &= \iint \tilde{P}_{\xi_1, \xi_2}^t(\psi_1, \psi_2) F_{x_1, x_2}^t(g_1, g_2) d^3g_1 d^3g_2, \end{aligned} \quad (13)$$

and that equivalent to (6) is

$$\tilde{P}_{\xi_1, \xi_2}^t(\psi_1, \psi_2) = \tilde{P}_{\xi_1, \xi_2}^0(2^{-t}\psi_1, 2^{-t}\psi_2) \left| \frac{\partial(2^{-t}\psi_1, 2^{-t}\psi_2)}{\partial(\psi_1, \psi_2)} \right| \quad (14)$$

where the Jacobian in (14) reduces to

$$[(1 - Ct\psi_1)(1 - Ct\psi_2)]^{-2}$$

Under the conditions that Γ is initially homogeneous and pairwise independent for all points,

$$\tilde{P}_{\xi_1, \xi_2}^0(\psi_1, \psi_2) = P^0(\psi_1) P^0(\psi_2)$$

and (14) becomes

$$\tilde{P}_{\xi_1, \xi_2}^t(\psi_1, \psi_2) = \tilde{P}^t(\psi_1) \tilde{P}^t(\psi_2) \quad (15)$$

where \tilde{P}^t is given by (10). With the substitution of (15) into (13), F_{x_1, x_2}^t integrates out and we obtain, using (12)

$$P_{x_1, x_2}^t(\psi_1, \psi_2) = P^t(\psi_1) P^t(\psi_2) \quad (16)$$

independent of the motion and of the points x_1 and x_2 . Consequently, homogeneity and pairwise independence is preserved. Results analogous to (16) also hold for multipoint pdf's of any order.

This invariance may be called a "white noise" invariance, as it implies for $D = 0$ that if the truncated power spectrum of Γ (in k -space) is initially flat, it will remain so, with all spectral components decaying at the same rate. Hence, once spatial correlations are lost, in the absence of diffusion they may not be regained. It would also be desirable, although we will not attempt this here, to be able to demonstrate with the continuity equation for $P^t[\psi | u]$ that Γ tends towards this equipartition.

The DI approximation evidently does not satisfy this invariance, at least one cannot tell by examination of Eqs. (1.11) and (1.12) of Ref. 6. However, the LHDII approximation (and also the abridged version) does preserve it, as one can tell from Ref. 7 by using Eqs. (38), (39), (45) - (47), the properties of

G when $D = 0$, and by noting that a property similar to Eq. (62) holds for the Fourier transform of the covariance of the generalized concentration field (normalized by $(2\pi)^{-3} \times$ volume of wavenumber space).

VI. THE USE OF SPECIFIC VOLUME

Let us turn our attention to the reciprocal of the concentration of the contaminant Γ , its specific volume

$$\Upsilon(x, t) = \Gamma^{-1}(x, t). \quad (17)$$

Any moment of Υ depends on all the moments of Γ , and vice versa. From (1) the behavior of Υ is given by

$$\mathcal{N}(x, t) \Upsilon(x, t) = C - \frac{2D}{\Upsilon(x, t)} |\nabla \Upsilon(x, t)|^2. \quad (18)$$

The quadratic nonlinearity of the reaction term in (1) has been replaced at this level in (18) by a logarithmic nonlinearity in the dissipation term. Such a nonlinearity is actually present in the single point hierarchy equations for either (1) or (18), as we have

$$\mathcal{N} \Gamma^n = -n C \Gamma^{n+1} - (n-1) D (\nabla \log \Gamma) \cdot (\nabla \Gamma^n)$$

$$\mathcal{N} \Upsilon^n = n C \Upsilon^{n-1} - (n+1) D (\nabla \log \Upsilon) \cdot (\nabla \Upsilon^n).$$

The standard practice (nonreactive passive scalar problem) for avoiding this dissipative term is to consider only the Γ hierarchy for multiple point correlations. After closure, certain points are made to coincide before substitution into lower order equations. The same approach could be used for $C \neq 0$, but one must be careful with the role played by the velocity field in the multipoint moments so that the O'Brien invariance is retained when the points coincide and $D = 0$. Unfortunately, the log term appears in the first equation of the Υ hierarchy. It is not apparent how to avoid dealing with it.

Because the dissipation term in (18) disappears when $D = 0$, whatever type of statistical approximation we apply to this term cannot destroy O'Brien's invariance. For $D = 0$ in homogeneous turbulence, (1) and (18) give the following hierarchies of moment equations

$$\begin{array}{ll} \dot{\Theta}_1 = -C\Theta_2 & \dot{\Phi}_1 = C \\ \dots & \dots \\ & \text{and} \\ \dot{\Theta}_n = -nC\Theta_{n+1} & \dot{\Phi}_n = nC\Phi_{n-1} \\ \dots & \dots \end{array}$$

where $\Theta_n = \langle \Gamma^n \rangle$ and $\Phi_n = \langle \Upsilon^n \rangle$ are averages defined over ensembles of initial data and velocity fields. Each hierarchy preserves O'Brien's invariance. However, the Φ hierarchy has the advantage of being closed at each level, and truncation does not alter from the true values the growth of specific volume or its moments, contrary to the Θ hierarchy. Another advantage of dealing with (18) is that the conditions under which the model (1) is at all applicable, dilute liquid

phase reactions for which D is extremely small and Υ is large, may allow the dissipation term to be linearized.

A substitution similar to (17) can be used for irreversible reactions of arbitrary order $N \neq 1$, $\Upsilon = \Gamma^{1-N}$, yielding an equation equivalent to (18) for the $(N - 1)$ st power of the specific volume.

VII. CONCLUSIONS

We have suggested two approaches that could be followed to guarantee preservation of O'Brien's invariance and the correct rates of decay of single point moments when $D = 0$. Description by the distribution function approach is difficult, and we have not pursued it far enough to demonstrate practical use. The use of specific volume in place of concentration in the moment approach is simpler, but the experimentalist would prefer to see results in terms of concentration, and the singularity in Υ for large times may be difficult to handle.

The generalizations (12) and (16) of the zero diffusivity invariance for the one and two point pdf's of Γ should, in retrospect, seem obvious on physical grounds. These properties should provide tests for the adequacies of either moment or distribution function approaches to either (1) or (18).¹⁴

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14. Sections III-V could just as well have been applied to $\Upsilon(x, t)$. For example, the equivalent of (7) for the pdf $P^t(\varphi)$ of Υ is $P^t(\varphi) = P^0(\varphi - Ct)$.